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Spectroscopy of individual LH2 complexes of *Rhodopseudomonas acidophila*: localized excitations in the B800 band

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Abstract

Using single-molecule detection techniques, fluorescence-excitation spectra from the B800 absorption band of individual light-harvesting complexes (LH2) of purple bacteria at 1.2 K were recorded. The spectra show distributions of narrow absorption lines that originate from excitations that are mainly localized on individual B800 bacteriochlorophyll *a* molecules. Spectroscopic details normally hidden under the inhomogeneous broadening, such as the degree of disorder in the pigment rings, the excited state dynamics and the energy transfer can be obtained.

1. Introduction

The initial steps of photosynthesis involve the transfer of energy of absorbed light to the photochemical reaction centre, where a charge separation takes place and the excitation energy becomes available in the form of chemical energy. Pigment–protein complexes surrounding the reaction centre function as highly efficient collectors of light that greatly increase the total absorption cross-section of the reaction centre, hence the name *light-harvesting* complexes. Of the known photosynthetic systems, the photosynthetic unit of purple bacteria is the most studied and best characterized. It consists of three types of pigment–protein complexes. The reaction centre (RC), where the charge separation takes place, is situated inside the light-harvesting 1 (LH1) complex [1]. Surrounding the LH1 complex, there are several light-harvesting 2 (LH2) complexes whose sole purpose is the absorption of light and the transfer of the energy towards the LH1 complex [2]. The elucidation of the high-resolution X-ray structure of the LH2 complex of *Rhodopseudomonas acidophila* [3], along with the lower-resolution structural information for LH1 [4], showed a remarkable symmetry in the arrangement of the light-absorbing pigments in these complexes. The LH2 complex contains 27 bacteriochlorophyll *a* (BChl *a*) molecules and (presumably) 18 carotenoids, held tightly into place by the surrounding protein matrix. The arrangement of the pigments is clearly visible in Fig. 1, where only the BChl *a* molecules are shown for clarity. The BChls *a* are arranged in two rings, both with a

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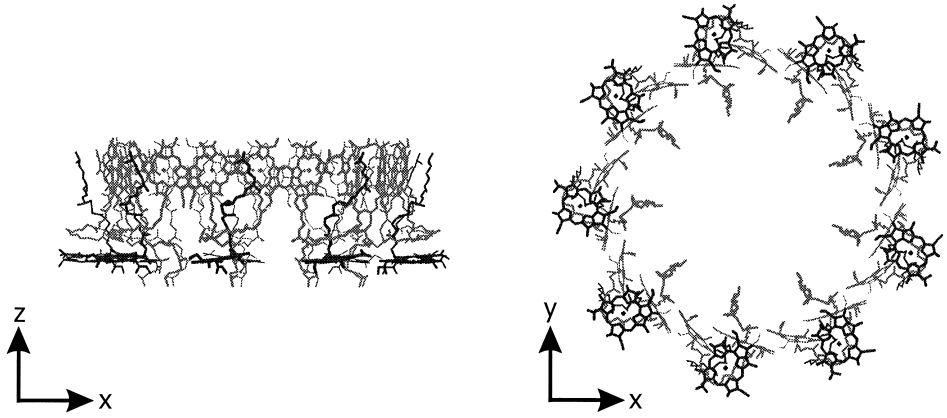

Fig. 1. Geometrical arrangement of the 27 BChl *a* molecules of the LH2 complex of *Rps. acidophila* obtained by X-ray crystallography. The data have been taken from the Brookhaven protein data bank (www.pdb.bnl.gov).

nine-fold symmetry axis that coincides with the cylindrical axis of the complex. One ring comprises 9 well-separated BChl *a* molecules with their molecular planes oriented perpendicular to the symmetry axis of the system. These molecules give rise to an absorption band lying around 800 nm (12 500 cm^{-1}) at room temperature, hence it is referred to as the B800 band. The other ring is constructed from the remaining 18 BChl *a* molecules, arranged as the blades of a turbine, with their molecular planes parallel to the 9-fold symmetry axis, and the corresponding absorption (B850) lies around 850 nm (11 750 cm^{-1}) at room temperature.

Upon excitation of one of the B800 molecules into the Q_y absorption (the lowest electronic transition of BChl *a*), rapid energy transfer is believed to occur between neighbouring B800 molecules [5–7] and between the B800 and the B850 band [6,8,9], both on a timescale of 1–2 ps. Owing to the smaller distances of the molecules in the B850 ring, the intermolecular interaction among these pigments is stronger as compared to that in the B800 assembly. Consequently, the energy is expected to be transferred more rapidly in the B850 manifold, on a timescale of $\sim$ 100 fs [10–12]. The energy is further transferred to the LH1 in 5–10 ps [13]. In the absence of LH1, fluorescent decay of the B850 molecules to the ground state occurs in $\sim$ 1 ns at a wavelength of 890 nm.

The observation of individual LH2 complexes was first reported by Bopp et al. [14]. At room temperature they measured fluorescence time traces, revealing the fluorescence lifetimes and photobleaching dynamics of the complexes. In a recent paper we demonstrated [15] that single-molecule detection techniques can also be applied to obtain the excitation spectrum of the B800 band of individual LH2 complexes at low temperature (1.2 K). The great advantage of this method is that we eliminate the inhomogeneous broadening of the absorption line caused by variations in the local environment of the pigments. In this paper we present the results of a more elaborate study of the B800 band of single LH2 complexes. We focus particularly on the question whether the excited states of the B800 band are completely localized on individual BChl *a* molecules or are delocalized over small groups of pigments. Also the consequences for the energy transfer mechanisms within the B800 manifold are discussed.

2. Experimental

Thin polymer films containing isolated LH2 were prepared by adding 1% polyvinyl alcohol (PVA; molecular weight 12 500) to a solution of 5×10^{-11} M LH2 from *Rps. acidophila* (strain 10 050) in buffer (0.1% LDAO/10 mM EDTA; pH 8.0) which was then spin coated on a LiF substrate. Using this method, polymer films with high optical quality and a thickness of < 1 μm could be produced.

After deposition of the film the substrate was mounted in a liquid-helium bath cryostat, together

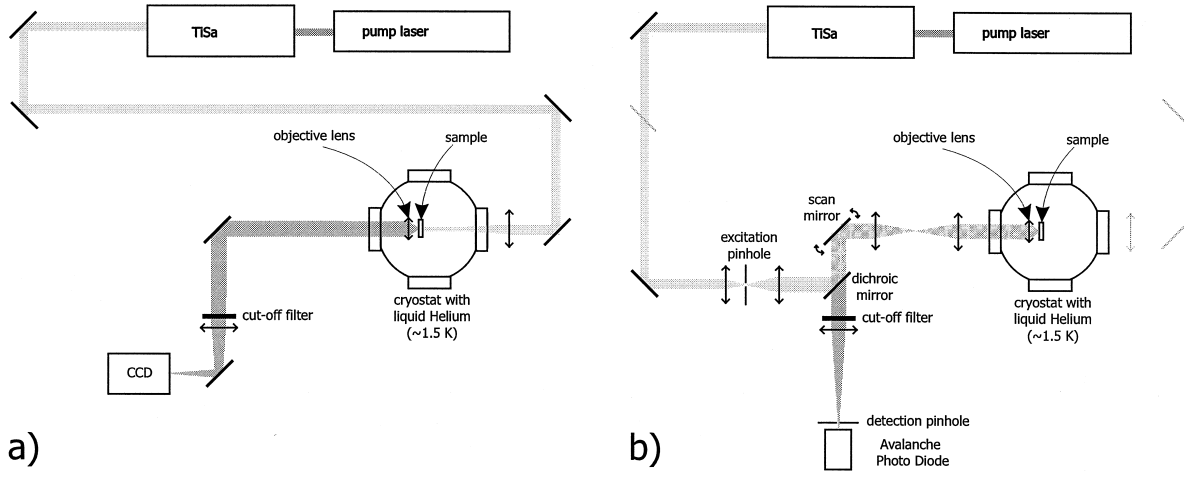

Fig. 2. Schematic representation of the experimental arrangement for the wide-field (a) and the confocal (b) experiments.

with an objective consisting of an aspheric singlet lens (N.A. = 0.55), and cooled to 1.2 K. The sample was excited by focusing the output of a cw, tunable Ti:sapphire laser (Spectra Physics, 3900 S) onto the sample (Fig. 2a) through a simple planoconvex lens with large focal length ($f = 250$ mm), creating a 100 μm large spot on the polymer film. The fluorescence emitted by the LH2 complexes is collected by the small objective lens present in the cryostat close to the sample and focused on a red-sensitive, back-illuminated CCD camera (Princeton Instruments, 512SB). Residual laser light is blocked by bandpass filters, which only transmit a 20 nm wide spectral window around 890 nm, the fluorescence wavelength of LH2. This selective fluorescence detection also strongly reduces the probability of observing features due to impurities. From the wide-field fluorescence images obtained by the CCD camera a spatially isolated LH2 complex can be selected for further spectroscopic investigation. In particular, a fluorescence-excitation spectrum of a specific complex could be obtained in the following two ways. A direct manner is by monitoring the fluorescence count rate on a small area in the CCD image, containing the diffraction-limited image of a single LH2 complex, as a function of the excitation wavelength. For reasons to be discussed in the next section, it appeared more advantageous to record the fluorescence-excitation spectra using a confocal detection scheme (Fig. 2b). After acquiring a wide-field image, the microscope was switched to the confocal mode by simply rearranging a mirror (Fig. 2b). From this point on the excitation light was directed towards the objective lens in the cryostat, illuminating a diffraction-limited excitation volume ($< 1\ \mu m^3$) of the sample. The scan mirror in the confocal microscope was then adjusted such that this excitation volume exactly coincides with the position of a particular LH2 complex found in the wide-field image. The fluorescence, collected by the same objective lens that illuminates the excitation spot, was focused on a confocally placed avalanche photodiode (EG&G, SPM 200), capable of single-photon counting. The presence of the detection pinhole ensures an extremely efficient suppression of the unwanted scattered light and fluorescence originating from areas in the sample outside the excitation volume. In the confocal detection mode the superior background suppression allows recording of fluorescence-excitation spectra with high signal-to-background ratios.

3. Results and discussion

In Fig. 3b a wide-field fluorescence image of a $50 \times 50\ \mu m^2$ region of the LH2-containing film is shown. While acquiring the image, the excitation wavelength is swept rapidly and repeatedly through a 50 cm^{-1} wide region in the centre part of the B800 absorption band to minimize any photo-induced

bleaching effects. The dark spots in the image correspond to the diffraction-limited images of individual LH2 complexes. This is evidenced by the following observations. The number of observed dots scales linearly with the LH2 concentration and agrees closely with the expected number of complexes estimated from the known concentration and sample thickness. For the experiments described in this paper the concentration of LH2 complexes was 50 pM, while the thickness of the film on the substrate is estimated to be $\sim 1\ \mu m$. This yields a probability of 3% to find exactly one complex in the confocal excitation volume. The probability to find two complexes in the same diffraction-limited volume element is already $< 0.1\%$.

The fluorescence-excitation spectrum of a particular LH2 complex can be recorded by reading out the total fluorescence signal of the corresponding small area on the CCD camera as a function of the excitation wavelength. This is illustrated in Fig. 3c. For comparison we show in Fig. 3a the spectrum of a large ensemble. The attractive feature of our technique is that it allows to register simultaneously the fluorescence-excitation spectra of all individual LH2 complexes within the CCD image. The disadvantage is that a large amount of background signal is present and that a high noise level is created by the on/off switching (blinking) of the fluorescence during the time that the excitation source is in resonance with one of the absorptions in the spectrum [15] (light-induced spectral diffusion). When reducing the speed of the wavelength scan and thus extending the time that the laser is in resonance with an absorption, the blinking behaviour is even enhanced, making it

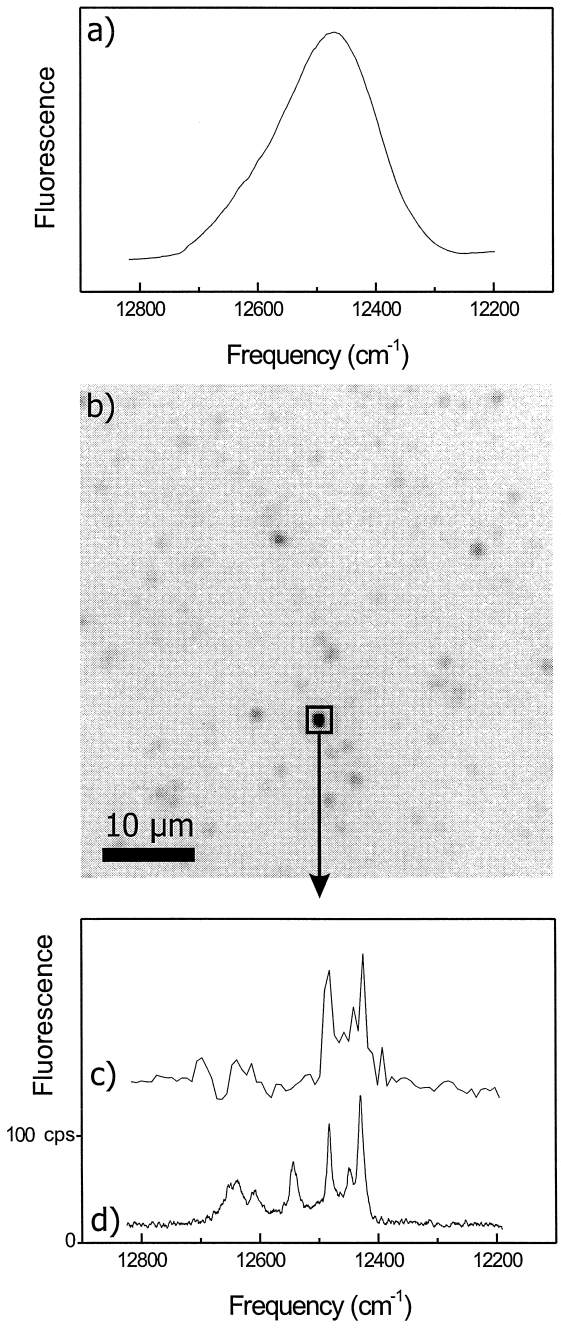

Fig. 3. (a) Fluorescence-excitation spectrum of the B800 band from a large ensemble of LH2 complexes from *Rps. acidophila*. (b) Image of a $50 \times 50\ \mu m^2$ sample region recorded in wide-field mode. The black dots represent diffraction-limited images of individual LH2 complexes from *Rps. acidophila*. The complex marked by the box has been selected for spectroscopy, see parts (c) and (d) of the figure. (c) B800 fluorescence-excitation spectrum of the single LH2 complex marked by the box in part (b) of the figure. The spectrum has been extracted from a sequence of wide-field images recorded while scanning the excitation wavelength. The total acquisition time amounted to 50 min and the illumination intensity was 80 W/cm^2. (d) B800 fluorescence-excitation spectrum of the same LH2 complex recorded in the confocal mode. The spectrum has been obtained by averaging in computer memory 200 independent fast scans of the excitation wavelength. The total acquisition time and illumination intensity were equal to that used in Fig. 2c. All experiments were performed at 1.2 K.

impossible to increase the signal-to-noise ratio by accumulation of the signal over a longer period of time. To overcome this problem, the microscope was switched to the confocal mode where the fluorescence is detected by an avalanche photodiode. In this way the complexes can only be studied one by one, but the superior background suppression and the capability to read out the avalanche photo diode much faster than the CCD camera allows us to record fluorescence-excitation spectra at the highest scan speed of the laser (3 nm/s). By accumulating many of such scans in the computer memory the spectrum of the B800 band of the same complex is obtained, and this result is shown in Fig. 3d. Clearly the fast sweeping of the excitation wavelength reduces considerably the blinking noise and spectra are obtained with a greatly enhanced signal-to-noise ratio. Additionally, this method opens the possibility to obtain information about the dynamics of the light-induced spectral-diffusion mechanisms.

For the 46 complexes studied, we observe a discrete pattern of typically 4–6 fluorescence-excitation lines with a spread of several nanometers around the centre of the ensemble absorption at 802 nm (12 470 cm^{-1}). To understand these spectra we consider the Hamiltonian describing the electronic structure of the B800 ring of nine interacting BChl *a* molecules which can be written as

$$H = \sum_{n=1}^{9} \left(E_y + \Delta_n\right) + \sum_{m \neq n} J_{mn}, \qquad (1)$$

where E_y denotes the energy of the Q_y transitions of the individual B800 molecules in a perfect, non-perturbed LH2 complex. The deviations from this value of the transition energies of the molecules, caused by inhomogeneities in the protein environment, often referred to as diagonal disorder, are indicated by Δ_n. The second term J_{mn} in the Hamiltonian is the interaction term between molecule *m* and molecule *n*. The relative values of Δ_n and J_{mn} determine whether the electronic excitation of the circular arrangement of the BChl *a* molecules corresponds to a coherent superposition of excited states or to localized excitations.

For $J \gg \Delta$ the excited states of the ring of pigments have to be described as coherent superpositions of the Q_y states of individual BChl *a* molecules. The eigenstates of the Hamiltonian (1) are Frenkel excitons given as:

$$|k\rangle = \frac{1}{\sqrt{N}} \sum_{n=1}^{N} e^{ikn\frac{2\pi}{N}} |n\rangle, \qquad (2)$$

where *N* denotes the total number of pigments in the ring and $|n\rangle$ describes the situation where molecule *n* is excited and the $N-1$ remaining molecules are in the ground state. For the B800 ring, this results in one nondegenerate state ($k = 0$) and four degenerate pairs of states ($k = \pm 1$, $k = \pm 2$, $k = \pm 3$, $k = \pm 4$) [16]. For negligible disorder, the lowest degenerate pair ($k = \pm 1$) holds almost all available dipole strength; the other transitions are essentially forbidden. Apparently this condition is not valid for the B800 system since we observe multiple, well-resolved peaks in the single-molecule spectrum.

The other extreme is $J \ll \Delta$, in which case the excitations are expected to be localized on one BChl *a* molecule [17]. In this case several absorption lines with a certain spread, reflecting the diagonal disorder Δ, are expected. To investigate the contention that the narrow lines in the fluorescence-excitation spectra correspond to excitations of individual BChl *a* molecules, polarization-dependent experiments were performed. In Fig. 4 seven fluorescence-excitation spectra of the B800 band of an individual LH2 complex are shown, each taken with a different orientation of the polarization vector of the exciting laser light. It is seen that the relative intensities of the absorption lines change appreciably upon changing the polarization. This is what one would expect if the excitations are localized on the individual BChl *a* molecules, because their Q_y transition dipole moments are arranged in a circular manner and, as a result of this, all have different orientations.

In this weak coupling limit, where $J \ll \Delta$, excitations may be assumed to be completely localized on individual pigments and energy transfer between B800 pigments can be described by Förster theory [7,18]. In this description the corresponding energy transfer rate is determined by the overlap of the fluorescence spectrum of the donor and the absorption spectrum of the acceptor. However, the absence of pronounced zero-phonon sidebands in our spectra shows that electron–phonon coupling is very weak. In view of the fact that the extremely narrow zero-

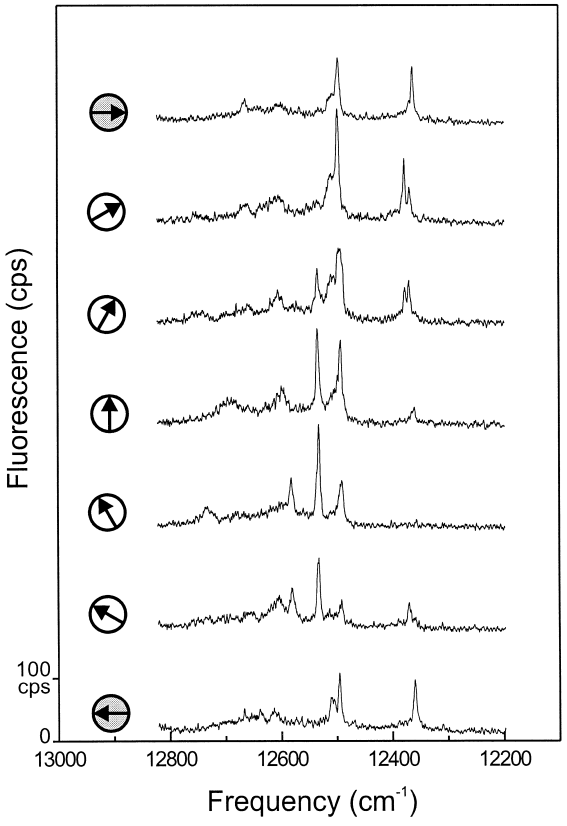

Fig. 4. Dependence of the B800 fluorescence-excitation spectrum of a single LH2 complex from *Rps. acidophila* on the polarization of the incident radiation. The polarization vector has been changed in steps of 30° from one spectrum to the next. The vertical scale is valid for the lowest trace, all other are displaced for clarity. The acquisition time for every trace was 25 min at an illumination intensity of 80 W/cm^2.

phonon lines are distributed throughout the complete B800 absorption it can be concluded that the spectral overlap of neighbouring B800 pigments is very small, in most cases negligible. Kolaczkowski et al. [19] have derived an analytical expression for energy transfer between pigments in the case of strong inhomogeneous broadening as a function of the electron–phonon coupling strength, and the energy difference in site energies. Using this expression, Wu et al. [9] showed that the Förster-overlap integral may indeed become very small when the electron–phonon coupling is weak. From an analysis of the spectra we estimate that the Huang–Rhys factor is < 0.3, while the average difference in site energy between nearest neighbors is 60 cm^{-1} as derived from the site energies in the fluorescence-excitation spectra of 46 individual LH2 complexes. Under these conditions Förster-type energy transfer would occur with a transfer time of tens of picoseconds. This result is in disagreement with conclusions from hole-burning experiments [5], which show that energy transfer among the B800 pigments at 1.2 K does occur at a much faster rate. From these measurements a transfer time of 0.9 ps for the most-blue absorbing pigments of the B800 band has been obtained, in agreement with the typical homogeneous linewidths in the single-molecule spectrum of the B800 band. We conclude that Förster-type energy transfer cannot explain these data and another mechanism must be responsible for energy transfer *within* the B800 band.

To further this point of view it is instructive to quantitatively examine the ratio $|J/\Delta|$ and to consider the validity of the assumption $J \ll \Delta$. In the point-dipole approximation J can be calculated by

$$J = \frac{1}{4\pi\varepsilon_0}\left(\frac{\vec{\mu}_1 \cdot \vec{\mu}_2}{r^3} - 3\frac{\left(\vec{\mu}_1 \cdot \vec{r}\right)\left(\vec{\mu}_2 \cdot \vec{r}\right)}{r^5}\right), \qquad (3)$$

where $\vec{\mu}_i$ is the transition dipole moment of molecule i and $\vec{r}$ is the intermolecular distance vector. Using $|\vec{\mu}| = 2.046 \times 10^{-29}$ C m (corresponding to 6.13 debye and a transition strength of $|\vec{\mu}^2| = 37.6$ debye^2) [20], $|\vec{r}| = 2.13$ nm, and the known geometry for the arrangement of the B800 molecules, one finds an interaction strength of -24 cm^{-1}. Using $\Delta = 130$ cm^{-1}, as determined from the average width of the distributions of site energies in the excitation spectra, one finds $|J/\Delta| \approx 0.2$. This ratio suggests that the diagonal disorder leads to a localization of the excitation mainly on one molecule with in addition a small density on 1 or 2 neighbouring pigments. We have verified this point of view by examining the polarization-dependent measurements from Fig. 4 in more detail. When combining all the polarization-dependent spectra of a single LH2 complex, 9 absorption lines should be observed, corresponding with completely localized excitations on the 9 individual BChl *a* molecules. However, in these combined spectra typically only 6–7 absorption lines were found, supporting the presence of a slight delocalization of the excited state over 2 or 3 neigh-

bouring BChl *a* molecules in the B800 manifold and a concomitant redistribution of oscillator strength.

Interestingly the ratio of $|J/\Delta|$ is similar to that observed in the case of the Fenna–Matthews–Olson (FMO) complex of green sulfur bacteria [21]. In the FMO complex, the value of Δ is dominated by the difference in site energies of the 7 inequivalent pigments in a subunit. The spectroscopic properties of the FMO complex at temperatures of 6 K and lower are described very well by a simple exciton model taking into account all the dipole–dipole interactions between the 7 pigments. More importantly, this model could also be applied to simulate the time-resolved absorbance difference spectra by incorporating linear electron–phonon coupling as described by Leegwater et al. [22]. This simulation was obtained with only one adjustable parameter, the absolute strength of the electron–phonon coupling. These results on the FMO complex show: (1) that exciton–phonon interactions are essential to describe the dynamical properties of the electronic excited states, even when $|J/\Delta|$ is as small as 0.2; (2) that in this case delocalization of the excitation is mostly limited to 2–3 pigments; and (3) that linear electron–phonon coupling can account quantitatively for relaxation between exciton states. The lifetimes of individual levels are determined by vibronic relaxation to the lower states in the exciton manifold of FMO, and vary between 100 fs and 2.5 ps, the shortest lifetimes being observed with excitation into the highest energy levels.

Since the values of $|J/\Delta|$ for the B800 pigments and the FMO complex are very similar, we suggest that the same conclusions may apply in the present case. The eigenstates of the B800 system at 1.2 K correspond to exciton states that are delocalized over a limited number of pigments, as proven by the polarization-dependent spectra from Fig. 4. At room temperature, strong mixing of the exciton states by vibronic coupling will occur, resulting in a further localization of the excitation [23,24]. The energy-transfer dynamics in the B800 band at 1.2 K can be explained by vibronic, radiationless relaxation between these exciton states, induced by linear exciton–phonon coupling.

In the preceding discussion of the dynamic properties of the excited states of the B800 ring we have ignored energy transfer to the B850 ring, a process that of course competes with relaxation within the excited state manifold of B800. The energy gap between B800 and B850 is $\sim 750\ cm^{-1}$, i.e. much larger than typical phonon frequencies. It is conceivable that energy transfer to B800 occurs via higher exciton states of the B850 pigment assembly which are near resonant with the B800 states [25,26]. It has been suggested by Schulten et al. [27] that this transfer is mediated by a Dexter-type process involving the carotenoids present in LH2. An experimental verification of the exact nature of this energy transfer mechanism deserves further attention and is currently in progress on a single pigment–protein level.

4. Conclusions

This study shows that excitations of the B800 band in LH2 of *Rhodopseudomonas acidophila* are localized on a limited number (1–3) of BChl *a* molecules. This contention is supported by polarization-dependent excitation spectra of the B800 band of individual LH2 complexes. Downward relaxation between exciton states, induced by linear electron–phonon coupling, is most likely responsible for the B800–B800 intraband energy transfer.

In view of the remarkable photostability of the system at low temperatures it is expected that a broad range of pigment–protein complexes can be spectroscopically investigated on the single-complex level. This will allow for a detailed study of the electronic structure of these pigment–protein complexes and of the interactions responsible for the efficient energy-transfer processes in the primary steps of photosynthesis.

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